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Final 2P

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A. Publications

The following reports of research supported by this grant were published in the past three years:

1 "The Photochemistry of Planetary Atmospheres and That of the Primitive Earth", J. P. Ferris, Advances in Organic Photochemistry" (A. Padwa, Ed.), Vol. 8, 1-65, 1987. (A review)

2. "HCN Formation on Jupiter by Photolysis of Ammonia-Acetylene Mixtures", Y. Ishikawa, H. Khwaja and J. P. Ferris, Origins of Life 16, 228 (1986).

3. "HCN and Chromophore Formation on Jupiter", J. P. Ferris and Y. Ishikawa, Nature, 326 777 (1987).

4. "The Formation of HCN and Acetylene Oligomers by Photolysis of Ammonia in the Presence of Acetylene: Applications to the Atmospheric Chemistry of Jupiter", J. P. Ferris and Y. Ishikawa, J. Am. Chem. Soc. 110, 4306 (1988).

5. "Photolysis of Ammonia in the Presence of Acetylene. A Plausible Explanation for the Formation of HCN and Chromophores on Jupiter", J. P. Ferris, Y. Ishikawa and K. Rahman, Origins of Life Evol. Biosphere 19, 489 (1989).

6. "The Photolysis of NH_3 in the Presence of Substituted Acetylenes: A Possible Source of HCN Oligomers and HCN on Jupiter", J. P. Ferris, R. R. Jacobson and J. C. Guillemin, submitted for publication.

Research supported by this grant was discussed in a presentation in the Goddard Scientific Colloquia Series, Greenbelt, MD, October 7, 1988 and at the NASA Summer School for Planetary Science at Caltech on August 14-18, 1989.

B. Summary Progress Report (Detailed data is given in the publications listed in Section III.A.)

1. HCN formation

We tested the theoretical proposal of Kaye and Strobel[11] that the HCN observed on Jupiter is the result of NH_3 photolysis in the presence of C_2H_2. Previous routes proposed for the formation of HCN on Jupiter by the coupled photochemistry of CH_4 and NH_3[12,13] or by the effect of shock waves or electric discharges[14] on CH_4 and NH_3 are not as plausible as the photochemical route from C_2H_2 and NH_3.[4] C_2H_2 and NH_3 occur together in the Jovian stratosphere so the reaction of radicals, formed by NH_3 photolysis, with C_2H_2 is a plausible Jovian scenario.

HCN is formed by photolysis of NH_3 in the presence of C_2H_2 at 298 K and 178 K in the presence and absence of excess hydrogen.[3] The yield of HCN is diminished ten-fold by lowering the temperature to 178 K due to the condensation of CH_3CN and

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$CH_3CH{=}NN{=}CHCH_3$, reaction intermediates in the formation of HCN, on the cell wall. No aziridine was detected as a reaction product.[11] Photolysis of $CH_3CH{=}NN{=}CHCH_3$ yields CH_3CN and the reaction with hydrogen atoms with CH_3CN yields HCN. From these and other data it was possible to propose a reaction pathway for HCN formation. The formation of HCN, CH_4 and C_2H_6 was confirmed by FT-IR analysis of the gaseous reaction products.

2. Chromophore formation

The hydrogen atoms produced by ammonia photolysis initiate the photochemical oligomerization of acetylene. This brown oligomer deposits as a very thin, transparent, yellow-brown film on the window of the quartz cell. This film is insoluble in traditional solvents and only dissolves in aqua regia. It was not possible to prepare a film thick enough to be removed from the cell because as the film increases in thickness it absorbs the light from the UV lamp and the NH_3 photolysis in the cell stops. It was possible to do FT-IR spectral analysis of the film by first performing the photolysis in a cell with CaF_2 windows which can be removed from the cell and placed in the FT-IR. The FT-IR spectrum revealed the presence of CH_3-, -CH_2-, and -NH- groupings. The spectrum obtained is different from that of polyacetylene indicating it has a quite different structure and much greater structural diversity.

The hydrogen atom-initiated polymerization of acetylene appears to be a plausible route to the formation of organic chromophores on Jupiter. The hydrogen atom-initiated reaction proceeds at 178 K and in the presence of 700 torr of hydrogen. The oligomer resulting from NH_3 photolysis incorporates -NH and -NH_2 groupings. It may be possible to generate other colors by the incorporation of -PH and -PH_2 groupings from PH_3.

This research demonstrated that the photolysis of NH_3 in the presence of C_2H_2 may provide insight into two persistent problems concerning Jupiter (1) the source of HCN (2) the source of the chromophores.